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14. ABSTRACT  After successful observation of gamma-emission from the 31-yr isomer of 178Hf induced by x-ray irradiation, the scientific interest for the triggering of long-lived nuclear spin isomers has greatly increased. We observed great efficiency of the triggering with low-energy (<20 keV) x-rays inducing the decay of 178m2Hf isomers that stresses all models, which might have been able to predict such an effect. Thus, there is a necessity to produce research lab-sized quantities of 178m2Hf isomers and in such a manner that the isomer samples could be used in research experiments as soon as possible. Since the production of 178m2Hf isomers will obviously be very expensive, careful and detailed analysis for the optimization of such production must be performed. The objective of this proposal is to find the most effective way of 178m2Hf isomer production for research uses and define the main parameters of nuclear facilities needed for that. Such research must minimize the risk of non-optimal production of 178m2Hf and can lead to significant cost savings.  As a result of such research, two or three spallation reactions, which would happen to be the most promising for effective production of small quantities of the 178m2Hf isomers, shall be chosen. For each of them all advantages and disadvantages will be listed. So one would be able to select, taking into account the available nuclear facilities and the accepted costs, the most suitable nuclear reaction needed to produce these small quantities of the 178m2Hf isomer. All the results of our research will be presented and discussed at scientific conferences and workshops and published in open scientific periodicals.					
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**and**

**EUROPEAN OFFICE OF AEROSPACE REASEARCH  
AND DEVELOPMENT**

**Kyiv - 2003**

# Final Report

## *Optimization of $^{178m2}\text{Hf}$ Isomer Production in Spallation Reactions*

### 1. Objectives

High-spin nuclear isomers and their interaction with external radiation fields were recognized as the most promising way to create a gamma-ray source controlled by soft x-ray devices. Unfortunately, there are only a few high-spin nuclear isomers the half-life of which is long enough to be useful for such research, namely  $^{180m}\text{Ta}$ ,  $^{178m2}\text{Hf}$  ( $T_{1/2} = 31$  y),  $^{179m}\text{Hf}$  ( $T_{1/2} = 24$  d) and  $^{177m}\text{Lu}$  ( $T_{1/2} = 160$  d). As for  $^{180m}\text{Ta}$ , though it is the only naturally existing nuclear isomer, its excitation energy is too low to produce a powerful source of pulsed gamma-ray radiation. On the other hand, while  $^{179m}\text{Hf}$  and  $^{177m}\text{Lu}$  isomers are rather long-lived, they turn out to be far from ideal for prediscvery experiments as well. Thus,  $^{178m2}\text{Hf}$  nuclear isomer looks as the only high-spin nuclear isomer attractive for such research. Really, having the excitation energy of isomeric state 2.45 MeV, 1 mg of  $^{178m2}\text{Hf}$  stores around 1 MJ of energy. Recent reports [1 - 10] have shown the principal possibility to trigger the release of such immense amount of energy over the time of the order of  $\mu\text{s}$  when an isomeric source was irradiated by low-energy x-rays. So in order to investigate the phenomenon in detail, just as to create and use such powerful sources of controlled gamma-ray radiation as effectively as possible, one needs to produce the  $^{178m2}\text{Hf}$  isomers in significant, at least lab-sized, quantities. Since the production of  $^{178m2}\text{Hf}$  isomers will obviously be very expensive, careful and detailed analysis for the optimization of such production must be performed and the main parameters of nuclear facilities needed for that must be defined.

According to available nuclear databases there were several successful attempts to produce  $^{178m2}\text{Hf}$  isomers in  $^{177}\text{Hf}$  ( $n, \gamma$ ),  $^{181}\text{Ta}$  ( $\gamma, p2n$ ),  $^{176}\text{Yb}$  ( $\alpha, 2n$ ) and  $^{181}\text{Ta}$  ( $p, \alpha$ ) reactions. As for ( $n, \gamma$ ) reaction, which looks quite straightforward, unfortunately, the cross-section of such neutron pumping of  $^{178m2}\text{Hf}$  is extremely small ( $\sim 2 \times 10^{-7}$  barn). Secondly, in order to produce the macroscopic samples of  $^{178m2}\text{Hf}$  one should carry out the mass-separation of isotopes, what means that the sources produced in such the way will be extremely expensive. On the other hand, ( $\gamma, p2n$ ) nuclear reaction is hardly very effective as well. So, the most effective nuclear reactions appear to be  $^{176}\text{Yb}$  ( $\alpha, 2n$ ) and  $^{181}\text{Ta}$  ( $p, \alpha$ ). At the same time, since the abundance of  $^{181}\text{Ta}$  is practically 100%, in the last case one can use the natural tantalum as well.

The biggest sample of  $^{178m2}\text{Hf}$  isomer (around 100  $\mu\text{g}$ ) was produced in spallation reaction using thick Ta targets and 800 MeV protons at the Los Alamos meson factory (LAMPF). In fact  $^{178m2}\text{Hf}$  isomer was accumulated as a sort of by-product and naturally the operation of LAMPF accelerator was not ever optimized for the best purity and highest yield production of the isomeric material [11]. As a result, first the irradiated samples were to be cooled down for a long time before they could be chemically processed. Secondly, practically all Hf isotopes, radioactive and stable, were accumulated in such spallation reaction as well, moreover most of them were produced in quantities orders of magnitude higher than  $^{178m2}\text{Hf}$  isomer. One of the radioactive Hf isotopes,  $^{172}\text{Hf}$  ( $T_{1/2} = 1.87$  years) creates very rich and intensive  $\gamma$ -ray background and, as a result,  $^{178m2}\text{Hf}$  isomeric sources produced in such the way can be used for any prediscvery experiments only after about 20 years cooling down. Naturally,  $^{172}\text{Hf}$  together with other Hf isotopes can be separated from  $^{178}\text{Hf}$  fraction by mass-separation techniques, but such process is rather difficult and expensive and used to result in

essential loss of the isomeric material [12]. The problem of separation of  $^{178g}\text{Hf}$  ground states and  $^{178m2}\text{Hf}$  isomeric states is not solved at all yet [13-14], though for some kinds of experiments and applications it could be quite crucial.

Much more qualitative  $^{178m2}\text{Hf}$  isomer samples were purposefully produced later at Dubna U-200 cyclotron using  $^{176}\text{Yb}$  ( $\alpha$ , 2n) reaction and 36 MeV  $\alpha$ -particles [15]. As a result, while the concentration of other Hf isotopes was really decreased significantly, the isomeric ratio simultaneously reached around 4%, much higher than in the case of the proton-induced spallation reaction at Ta. On the other hand, absolute productivity yield of this method turns out to be several orders of magnitude less compared to the spallation method due to much thinner targets and beam power removal problems.

At the same time, an attempt has been made to optimize the production of  $^{178m2}\text{Hf}$  isomers in the spallation reactions with protons [16]. Ta and Re targets were irradiated at Dubna synchrocyclotron at the energies of 100, 200 and 660 MeV and at the energies of 150, 300, 450 and 660 MeV, respectively. And although Re target did not justify hopes for higher purity and productivity of  $^{178m2}\text{Hf}$  isomer sources, none the less quite a lot of very interesting observations and conclusions have been drawn and some valuable recommendations how to improve the produced isomeric sources have been made as well. For instant, it has been illustrated and proved that  $^{178m2}\text{Hf}$  isomeric sources of the same quality or even better can be produced using rather low energetic protons (100-200 MeV).

On the other hand, there can be other quite perspective spallation reactions for  $^{178m2}\text{Hf}$  isomers production, maybe even using the heavy ion beams. So, in order to produce  $^{178m2}\text{Hf}$  isomers in lab-sized quantities and for the reasonable costs, it would be very important to perform very careful analysis of all the available possibilities.

The objective of the project was to analyze the  $^{178m2}\text{Hf}$  isomer production in different spallation reactions exploiting the STAPRE and ALICE code simulations and this objective has been subdivided into several tasks:

- Calculation of energy dependence for excitation functions. As a result, highest cross-sections of  $^{178m2}\text{Hf}$  isomer production in different spallation reactions have been estimated. Since nuclear reaction cross-section depends strongly on the energy of impact particle, in order to get the maximal isomer output one should define the optimal particle energy as well.
- Calculation of energy dependence for isomer ratios. In any nuclear reaction, along with  $^{178m2}\text{Hf}$  isomer,  $^{178g}\text{Hf}$  ground state would be produced also. So, for a given reaction the bigger  $^{178m2}\text{Hf}/^{178g}\text{Hf}$  ratio (termed the isomeric-to-ground or isomeric ratio) the better this reaction for  $^{178m2}\text{Hf}$  isomer production.
- Calculation of energy dependence for excitation functions of any nuclear reaction products that could be considered as undesirable admixtures.
- Selection of irradiation mode and estimate of costs for the isomer separation.

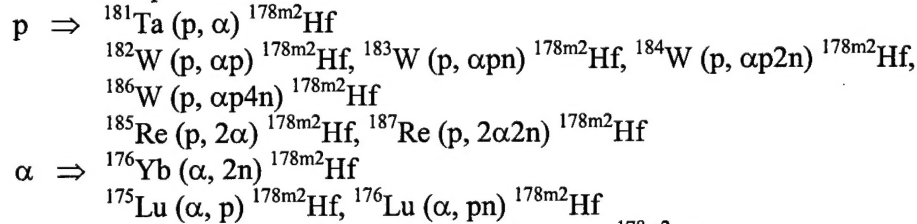
## **2. Qualitative comparison of spallation reactions with charged particles looking attractive for $^{178m2}\text{Hf}$ isomer production**

Along with (p,  $\alpha$ ) and ( $\alpha$ , 2n) reactions already used for  $^{178m2}\text{Hf}$  isomer production, any nuclear reaction with  $\alpha$ -particles like ( $\alpha$ , xn) or ( $\alpha$ , pxn) could be very promising for  $^{178m2}\text{Hf}$  isomer production as well [17 - 22]. As for the first type of nuclear reactions, in these reactions the energy of bombarding particles must exceed by 3-4 times the energy, at which the maximum of excitation function is observed. Investigations of ( $\alpha$ , xn) nuclear reactions have shown that at such energies the  $^{178m2}\text{Hf}/^{178g}\text{Hf}$  isomeric-to-ground state ratios are enhanced by 2-3 times and further rise of isomeric ratios at higher energies can be expected. Unfortunately, the heaviest stable Yb isotope is  $^{176}\text{Yb}$ , moreover  $^{175}\text{Yb}$  is not stable, so the

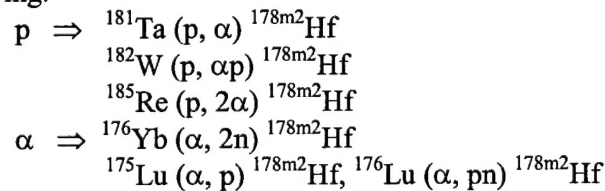
first class of discussed reactions can be presented by the only one already exploited for  $^{178m2}\text{Hf}$  isomer production.

As for the second type of nuclear reactions mentioned above like  $(\alpha, \text{pxn})$ , when a proton escapes the nucleus as well, it can be presented at least by two reactions -  $^{175}\text{Lu}(\alpha, \text{p})^{178m}\text{Hf}$  and  $^{176}\text{Lu}(\alpha, \text{pn})^{178m}\text{Hf}$ . The intensive study of isomeric ratios for such the reactions has shown that  $^{178m2}\text{Hf}/^{178g}\text{Hf}$  isomeric-to-ground state ratios increase rapidly above some threshold and then remain practically stable with the increase of bombarding particle energy. Such dependence allows selecting the irradiation mode when we could both maximize  $^{178m2}\text{Hf}$  yield and minimize amount of other Hf isotopes.

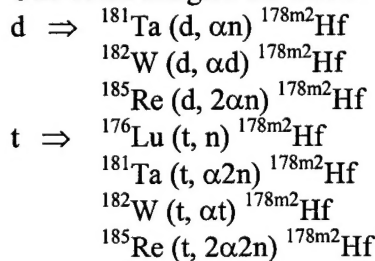
Thus, there is a number of nuclear reactions with charged particles looking attractive for  $^{178m2}\text{Hf}$  isomer production:



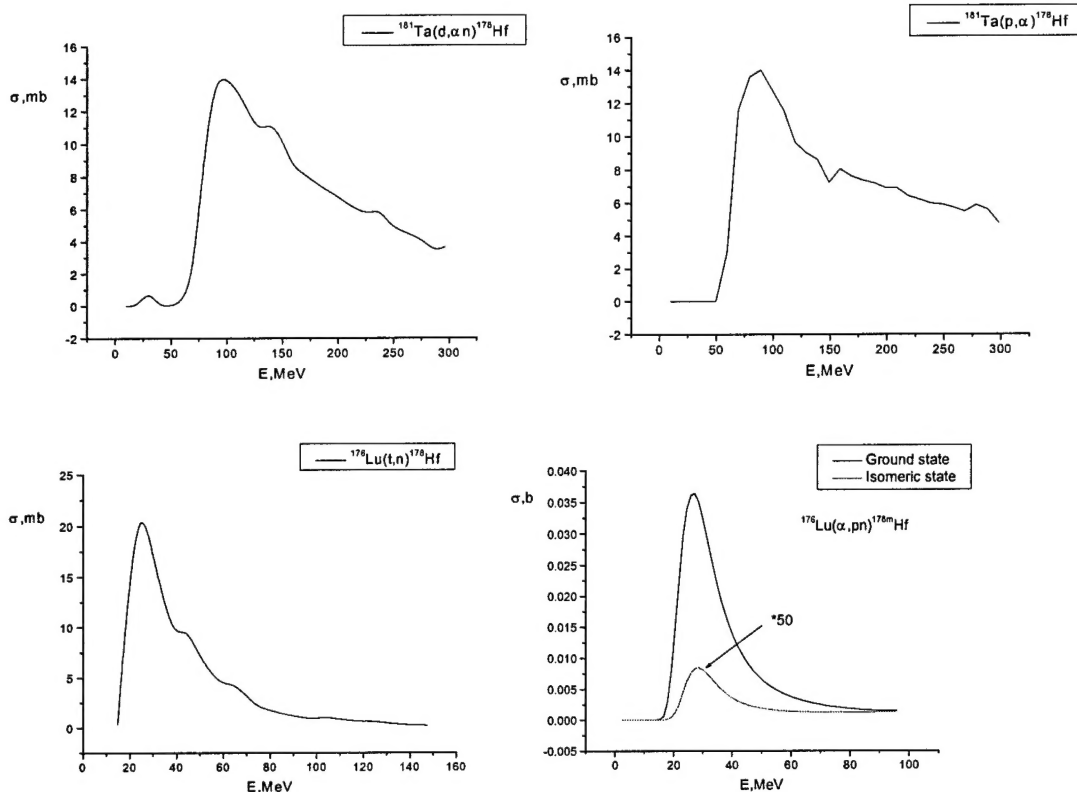
One can expect that the biggest cross-sections for  $^{178m2}\text{Hf}$  isomer production should be in such nuclear reactions, when the number of outgoing particles is small, since the bigger a number of outgoing particles, the higher a threshold of the reaction, moreover the higher an energy of bombarding particles, the bigger a number of competitive channels. On the other hand, when the number of emitted nucleons is small, the  $^{178m2}\text{Hf}/^{178g}\text{Hf}$  isomeric-to-ground state ratio is expected to be much less than unity, because of very high spin of the isomer (16) and the small transferred angular momentum. The increase of isomeric ratios at the bigger number of outgoing particles could slightly compensate the decrease of  $^{178m2}\text{Hf}$  yield due to the higher threshold and more numerous competitive channels of the nuclear reaction, though such compensation would hardly be large enough. So, the list of nuclear reactions with light charged particles that look as the most useful for  $^{178m2}\text{Hf}$  isomer production would be the following:



One could imagine the similar nuclear reactions with deuterons and tritons as well:



Anyway, the productivity of  $^{178m2}\text{Hf}$  isomer for such reactions is hardly higher, than for traditional bombarding particles such as protons and  $\alpha$ -particle. As a sort of test we have performed the ALICE calculations of  $^{178g}\text{Hf}$  production for one nuclear reaction with deuterons and one nuclear reaction with tritons and compared them to the analogous nuclear reactions with protons and  $\alpha$ -particles:



One can notice that in the case of  $^{176}\text{Lu}$  target,  $^{178g}\text{Hf}$  cross-section is about two times higher for  $^{176}\text{Lu}(\alpha, pn)^{178g}\text{Hf}$  reaction, than in the event of  $^{176}\text{Lu}(t, n)^{178g}\text{Hf}$ . In the case of  $^{181}\text{Ta}$  target, the  $^{178g}\text{Hf}$  cross-section turns out to be almost the same for both reactions -  $^{181}\text{Ta}(d, \alpha n)^{178g}\text{Hf}$  and  $^{181}\text{Ta}(p, \alpha)^{178g}\text{Hf}$ . It means that there is not any reason to use deuterons and tritons, since they present no advantage compared to protons and  $\alpha$ -particles.

As for the reactions with heavy ions, all the attractiveness of them for the production of such high spin nuclear isomers as  $^{178m2}\text{Hf}$  disappears when one takes into account the stopping power of heavy ions in the target material. For instant, even for the reactions with  $\alpha$ -particles compared to the ones when protons are used, while the cross-sections and isomeric ratios are higher, the productivity of  $^{178m2}\text{Hf}$  isomers happens to be still lower because of much more thin targets.

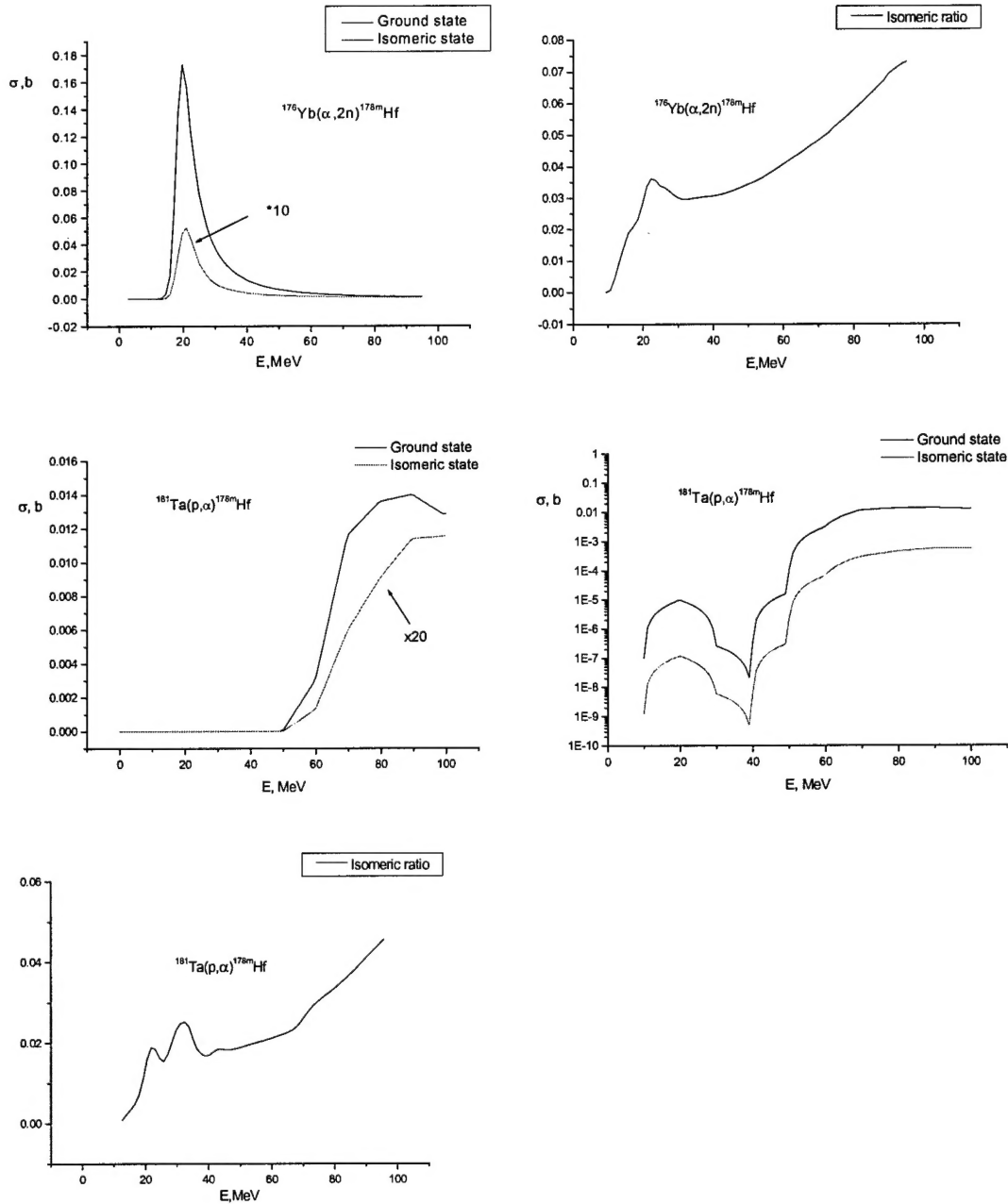
Moreover, all the nuclear reactions with heavy ions used to go through a compound nucleus and, as a result, the excitation energy of the latter appears to be not very high (quite close to the yrast band). It means that isomeric ratios for such reactions practically do not depend on the energy of bombarding heavy ions and this fact has been confirmed experimentally. At the same time, for many nuclear reactions with lighter particles some increase of isomeric ratios is still observed at the higher energies of projectiles.

Additional factor, that light nuclei have practically the same number of protons and neutrons, seems to exclude from consideration any nuclear reactions with bombarding ions heavier than  $\alpha$ -particle, since the less  $Z$  of the target the more  $p$ -deficient projectile one needs to reach  $^{178m2}\text{Hf}$  isomer. For example, while for  $^{169}\text{Tm}$  it should be something like  $(3p6n)$ , for  $^{159}\text{Tb}$  -  $(7p12n)$  and so on. So the most perspective charged projectiles for the  $^{178m2}\text{Hf}$  isomer production turn out to be protons and  $\alpha$ -particles and further consideration of ours will be restricted by that type of nuclear reactions.

### 3. STAPRE and ALICE code simulations of excitation functions and isomeric ratios for the most perspective spallation reactions with charged particles

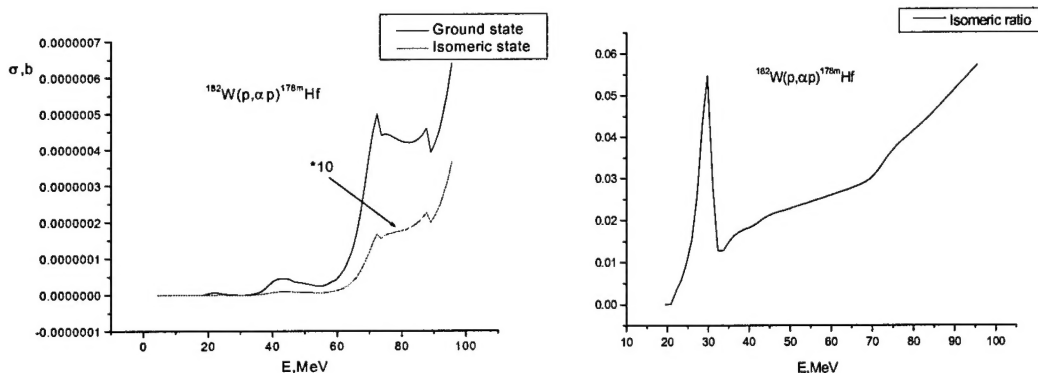
All calculations of energy dependence for excitation functions and isomeric ratios have been done on the basis of pre-equilibrium and cascade-evaporating models using the STAPRE and ALICE code simulations, furthermore while the STAPRE code was exploited to calculate the isomeric ratios, the ALICE code was exploited to simulate the excitation functions of respective ground states.

One could expect that the most promising spallation reactions for  $^{178m2}\text{Hf}$  isomer production would be  $^{176}\text{Yb}(\alpha, 2n)^{178m2}\text{Hf}$  and  $^{181}\text{Ta}(p, \alpha)^{178m2}\text{Hf}$  due to the biggest cross-sections of  $^{178g}\text{Hf}$  ground state production and not bad isomeric ratios compared to other competitive reactions from the list.



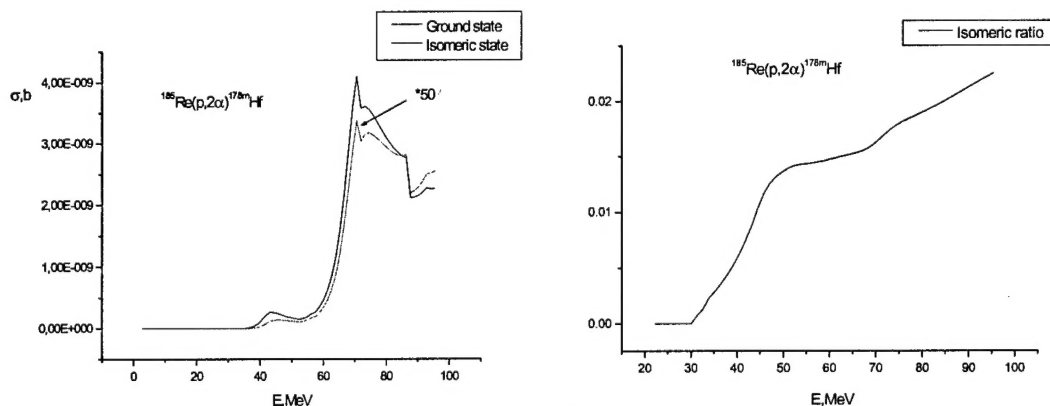


All the same, reactions  $^{182}\text{W} (p, \alpha p) ^{178m2}\text{Hf}$ ,  $^{185}\text{Re} (p, 2\alpha) ^{178m2}\text{Hf}$ ,  $^{175}\text{Lu} (\alpha, p) ^{178m2}\text{Hf}$  and  $^{176}\text{Lu} (\alpha, pn) ^{178m2}\text{Hf}$  look quite attractive as well. As for  $^{182}\text{W} (p, \alpha p) ^{178m2}\text{Hf}$ , one could hardly expect that it would be better, than  $^{181}\text{Ta} (p, \alpha) ^{178m2}\text{Hf}$ . Really, although the isomeric ratios turn out to be practically the same (up to 0.06), the mean cross-section happens to be at least an order of magnitude less.



As for  $^{185}\text{Re} (p, 2\alpha) ^{178m2}\text{Hf}$ , the point is that in the case of Re spallation with high-energy protons nuclei with mass  $A = 178$  happen to be located near the maximum of mass distribution. So, in the case of Re targets the expectation was that higher angular momentum should be produced for  $A=178$  isobars compared to the case of Ta targets and, as a result, the isomeric ratio  $^{178m2}\text{Hf}/^{178g}\text{Hf}$  would be higher. However, since  $^{178}\text{Hf}$  is located at the tail of isobaric charge distribution, its yield turns out to be orders of magnitude lower than the total yield of  $A = 178$  isobars. Unfortunately,  $^{178m2}\text{Hf}$  isomers result only as an independent yield, i.e. produced directly in the reaction, so their number should be very small, while the cumulative production of  $^{178g}\text{Hf}$  ground state, in other words resulted from the decay of heavier unstable isobars, would be large. It means that in the event of  $^{185}\text{Re} (p, 2\alpha) ^{178m2}\text{Hf}$  reaction there are even two disadvantages:

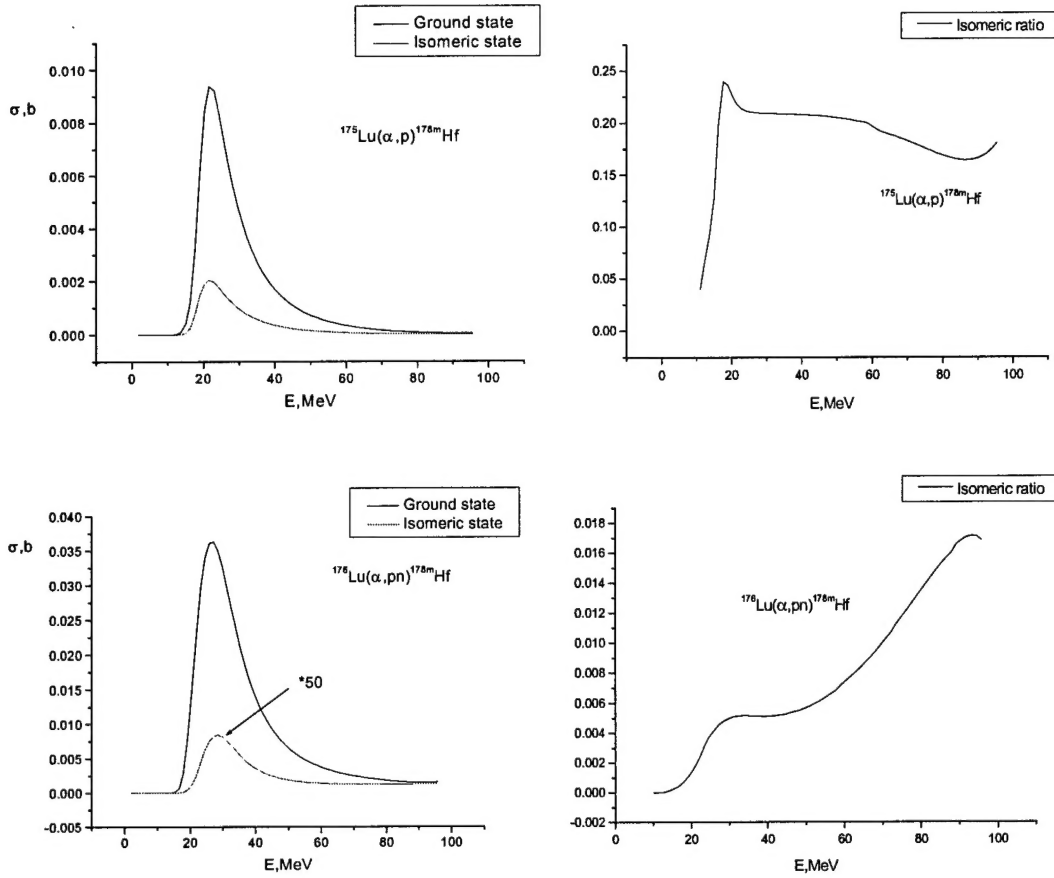
- $^{178m2}\text{Hf}$  isomer yield is very small;
  - isomeric ratio  $^{178m2}\text{Hf}/^{178g}\text{Hf}$  is small as well.
- and it was confirmed experimentally [16].



As for  $^{176}\text{Lu} (\alpha, pn) ^{178m2}\text{Hf}$  reaction the mean cross-sections for  $^{178m2}\text{Hf}$  isomer production and isomeric ratios are much lower, than in the event of  $^{176}\text{Yb} (\alpha, 2n) ^{178m2}\text{Hf}$  reaction. In the case of  $^{175}\text{Lu} (\alpha, p) ^{178m2}\text{Hf}$  reaction, though the mean cross-sections for  $^{178m2}\text{Hf}$  isomer production happen also to be much lower, than in the event of  $^{176}\text{Yb} (\alpha, 2n)$



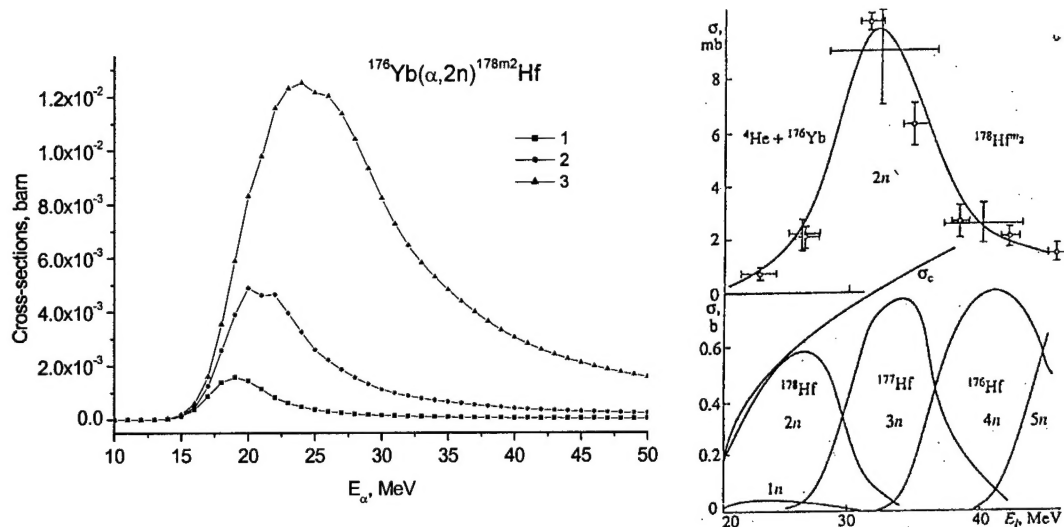
$^{178m2}\text{Hf}$  reaction, the isomeric ratios seem to be a little higher. It means that  $^{178m2}\text{Hf}$  isomer productivity of  $^{176}\text{Yb}(\alpha, 2n)^{178m2}\text{Hf}$  reaction should be higher than the productivity of  $^{175}\text{Lu}(\alpha, p)^{178m2}\text{Hf}$  reaction and the productivity of latter - higher than the productivity of  $^{176}\text{Lu}(\alpha, pn)^{178m2}\text{Hf}$ .



In order to be sure that all the conclusions are correct, one should check or calibrate the STAPRE and ALICE codes using well-known theoretical or precise experimental results. The point is that there are:

- difficulties in calculating the angular momentum (l-distribution) for reaction residues;
- uncertainties in the prediction of excitation functions and isomeric ratios even for known l-distribution because of the sensitivity of calculation results to the choice of numerical parameters.

Just as an example how the choice of numerical parameters influences upon the results of calculations, below one can see the cross-sections for  $^{176}\text{Yb}(\alpha, 2n)^{178m2}\text{Hf}$  reaction calculated using the STAPRE code with different sets of partial waves for p, n and  $\alpha$ -particle (3,3,6), (5,5,10) and (7,7,14) that stand for 1, 2 and 3 respectively. For the comparison we have presented the measured  $^{178m2}\text{Hf}$  cross-sections (all the stable isotope  $^{176}, ^{177}, ^{178}\text{Hf}$  excitation functions were calculated using the statistical model code) [23].



Thus, the third set of partial waves for p, n and  $\alpha$ -particle (7,7,14) looks as the most proper one for this reaction, at least much better than the first (3,3,6) and the second (5,5,10) sets.

Quite recently new experimental data have been obtained for  $^{181}\text{Ta} (p, \alpha) ^{178\text{m}2}\text{Hf}$  reaction [16] that would be ideal to calibrate the STAPRE code at least up to 100 MeV. However, in order to use the mentioned above experimental results one should take into account that the bombarding particle loses its energy while passing through the target material. For the target of known thickness the yield,  $Y$  for a reaction product per one projectile is defined as:

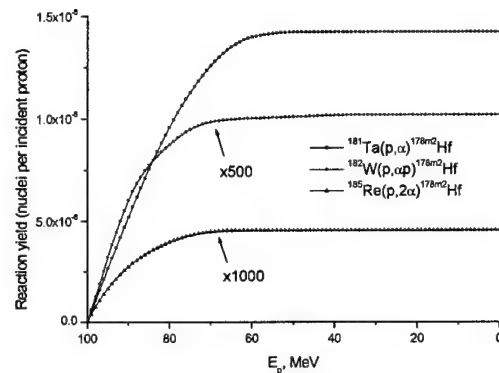
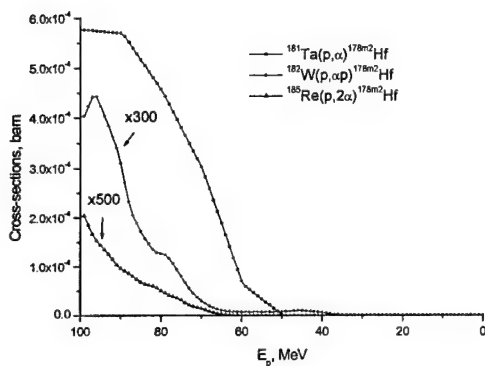
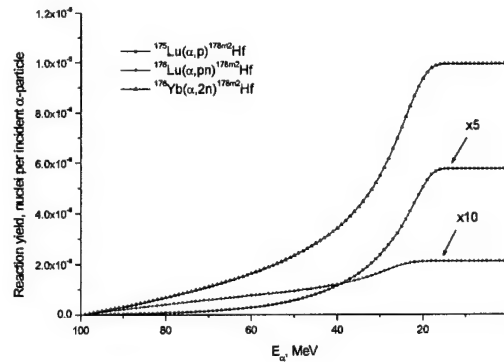
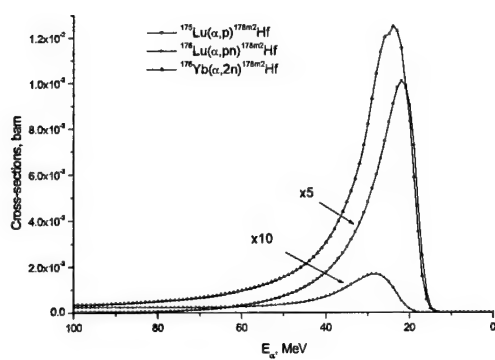
$$Y = \int_{E_{\min}}^{E_{\max}} \sigma(E) \left( \frac{dE}{dx} \right)^{-1} dE,$$

where  $E_{\max}$  and  $E_{\min}$  stand for the bombarding particle energy range in a target of known thickness and  $dE/dx$  is the energy dependent stopping power of the projectile in the target material (expressed in  $\text{MeV}/\text{atom} \cdot \text{cm}^2$ , if  $E$  is in MeV and  $\sigma$  - in  $\text{cm}^2$ ). The mean cross-section is connected with the yield through the expression:

$$\bar{\sigma} = Y \left( \int_{E_{\min}}^{E_{\max}} \left( \frac{dE}{dx} \right)^{-1} dE \right)^{-1}.$$

Having performed such quite simple calculations, we carefully checked both STAPRE and ALICE codes.  $^{178\text{m}2}\text{Hf}$  mean cross-section obtained in  $^{181}\text{Ta} (p, \alpha) ^{178\text{m}2}\text{Hf}$  reaction was used for the STAPRE code calibration. In the case of ALICE code, we used both  $^{172}\text{Hf}$  and  $^{178}\text{W}$  mean cross-sections for calibration of this reaction. As a result of such calibration procedure, the most proper angular momentum distributions for the reaction residues have been identified. Thus, both the excitation functions and the isomeric ratios calculated by the STAPRE and ALICE codes should be rather correct.

At the same time, discussing all the mentioned above spallation reactions, we should take into account not only cross-sections and isomeric ratios, but the productivity of each reaction as well.

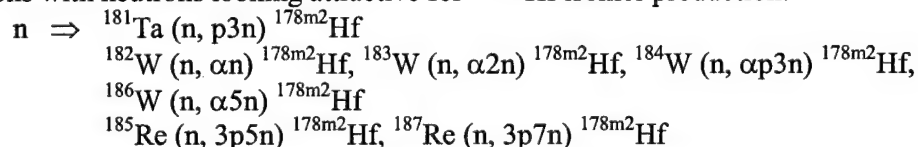


Thus, as for nuclear reactions with  $\alpha$ -particles,  $^{176}\text{Yb}(\alpha, 2n)^{178m2}\text{Hf}$  looks as the most attractive one, at the same time  $^{175}\text{Lu}(\alpha, p)^{178m2}\text{Hf}$  and  $^{176}\text{Lu}(\alpha, pn)^{178m2}\text{Hf}$  are about 10 and 50 times worse, respectively. As for nuclear reactions with protons,  $^{181}\text{Ta}(p, \alpha)^{178m}\text{Hf}$  reaction looks as the most attractive one, at the same time  $^{182}\text{W}(p, \alpha p)^{178m}\text{Hf}$  and  $^{185}\text{Re}(p, 2\alpha)^{178m}\text{Hf}$  are about 800 and 3000 times worse, respectively.

#### 4. STAPRE and ALICE code simulations of excitation functions and isomeric ratios for the most perspective spallation reactions with neutrons

Another kind of nuclear reactions that could also be very interesting - reactions with neutrons. And although in such reactions one can hardly expect so high cross-sections and isomeric ratios as in the case of reactions with charged particles, neutrons as bombarding particles have several distinguished advantages. First, neutrons are very penetrative particles, so one can use very massive targets. Secondly, there are a great variety of different neutron sources quite powerful and relatively not very expensive, including nuclear reactors.

In full analogy to nuclear reactions with protons, there is a number of nuclear reactions with neutrons looking attractive for  $^{178m2}\text{Hf}$  isomer production:



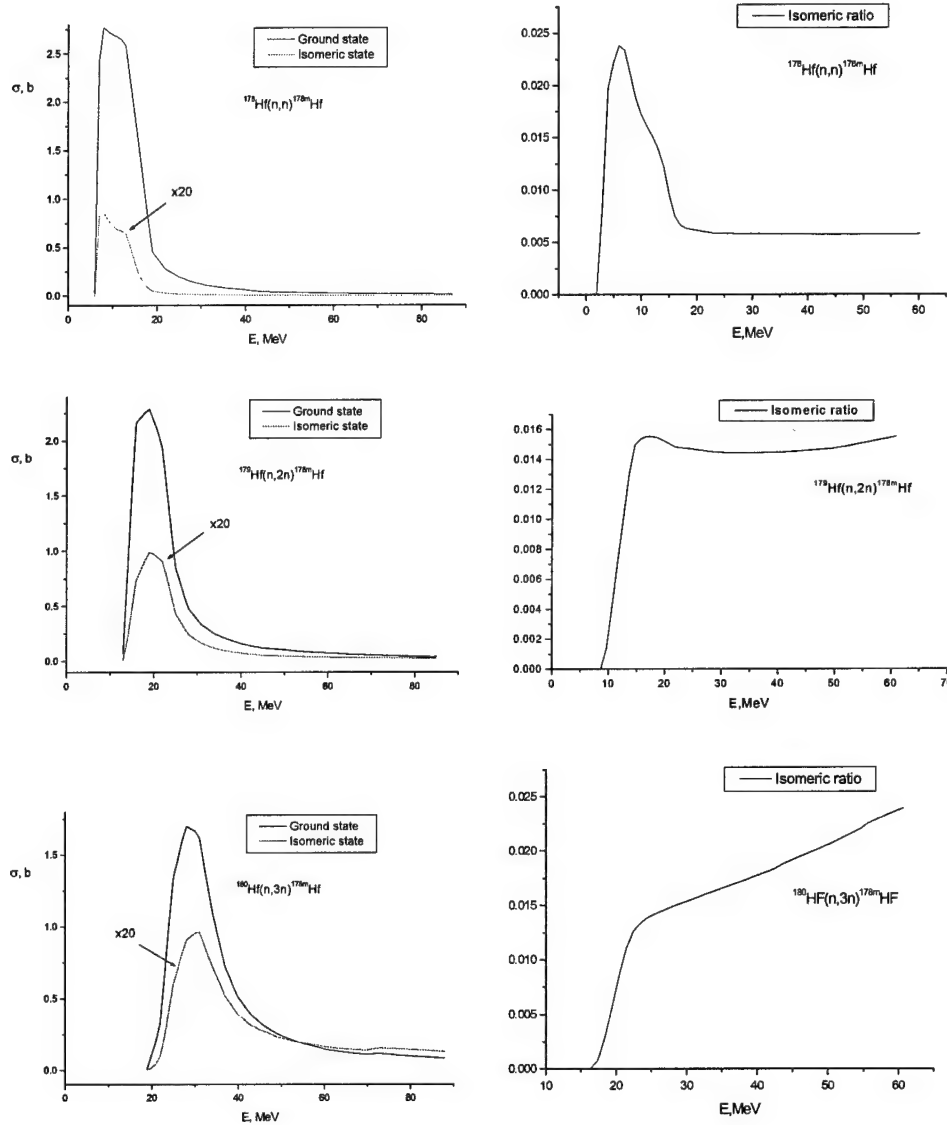
For the same reasons as in the case of nuclear reactions with protons we expect that nuclear reaction  $^{182}\text{W}(n, \alpha n)^{178m2}\text{Hf}$  should be the most perspective.

At the same time, using nuclear reactors for the production of  $^{178m}\text{Hf}$  isomers one should take into account that energy distribution of reactor neutrons is defined as:

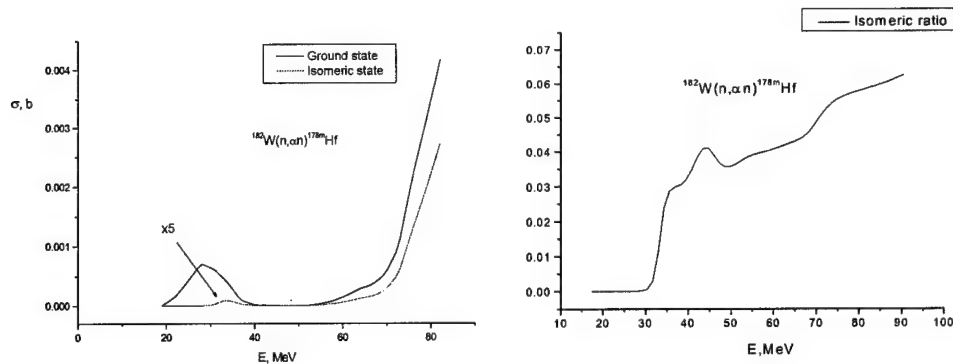
$$N(E) \sim e^{-E} \times sh[(2E)^{1/2}]$$

and has specific form with maximum around 1 MeV.

So we tried to calibrate the STAPRE code for the reactions with reactor neutrons as well using experimental data obtained quite recently for a number of highly enriched Hf targets [24]. Isomeric ratios have been taken from the STAPRE calculations and the excitation function for  $^{178g}\text{Hf}$  - from the ALICE calculations.



After that we have calculated the productivity of nuclear reaction  $^{182}\text{W}(n, \alpha n)^{178m2}\text{Hf}$ , the obvious advantage of which compared to the reactions with Hf targets is that no mass-separation is needed to obtain  $^{178m2}\text{Hf}$  isomeric source.



As a result, the mean cross-sections for mentioned above nuclear reactions with reactor neutrons are presented below (Table1).

Table1. Mean cross-sections of  $^{178\text{m}}\text{Hf}$  isomer and  $^{178\text{g}}\text{Hf}$  ground state for nuclear reactions with reactor neutrons.

Nuclear reaction	$\sigma_{\text{m2}}, \text{ b}$	$\sigma_{\text{g}}, \text{ b}$
$^{178}\text{Hf}(n, n')$	$7.369 \cdot 10^{-4}$	$4.626 \cdot 10^{-2}$
$^{179}\text{Hf}(n, 2n)$	$1.455 \cdot 10^{-6}$	$9.785 \cdot 10^{-5}$
$^{180}\text{Hf}(n, 3n)$	$1.510 \cdot 10^{-9}$	$1.159 \cdot 10^{-7}$
$^{182}\text{W}(n, \alpha n)$	$2.249 \cdot 10^{-15}$	$5.269 \cdot 10^{-11}$

Comparing the obtained results for nuclear reactions with reactor neutrons and the results of calculations for nuclear reactions with charged particles, one can note that really the mean  $^{178\text{m}}\text{Hf}$  and  $^{178\text{g}}\text{Hf}$  cross-sections for  $^{182}\text{W}(n, \alpha n)^{178\text{m}}\text{Hf}$  reaction look still far from the values needed to consider this nuclear reaction as perspective for  $^{178\text{m}}\text{Hf}$  isomer production. On the other hand, since the results of our calculations contradict significantly to the experimental data obtained recently for a number of targets of highly enriched Hf isotopes [24], it would be very interesting to check the obtained results for nuclear reaction  $^{182}\text{W}(n, \alpha n)^{178\text{m}}\text{Hf}$  with reactor neutrons both theoretically and experimentally.

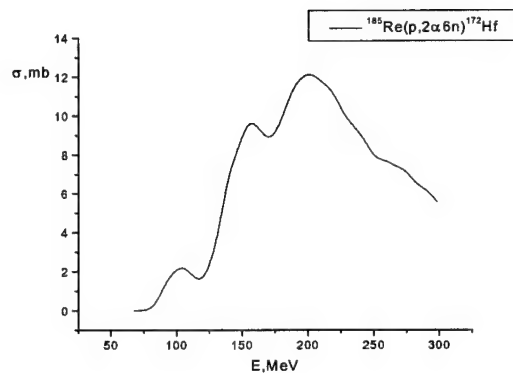
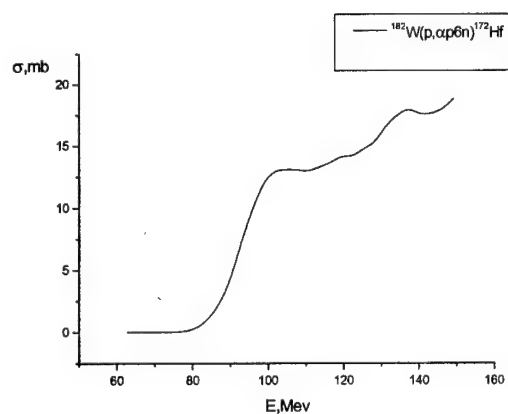
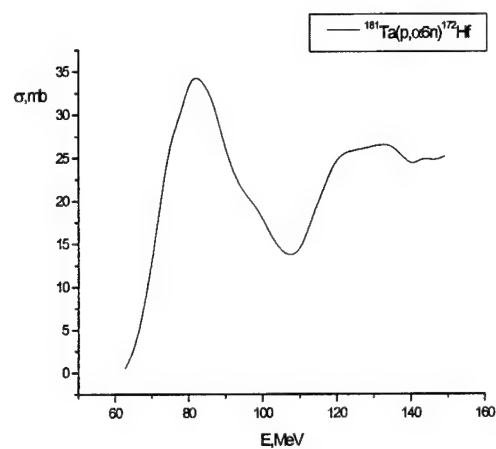
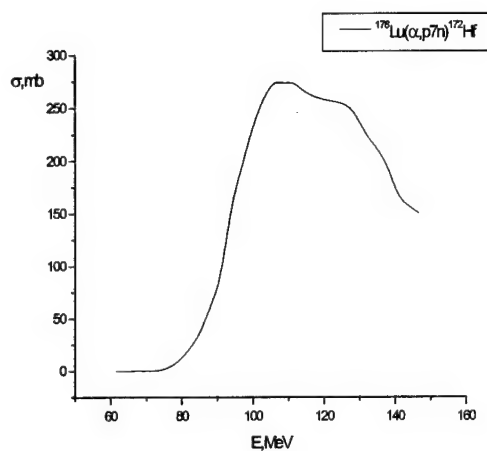
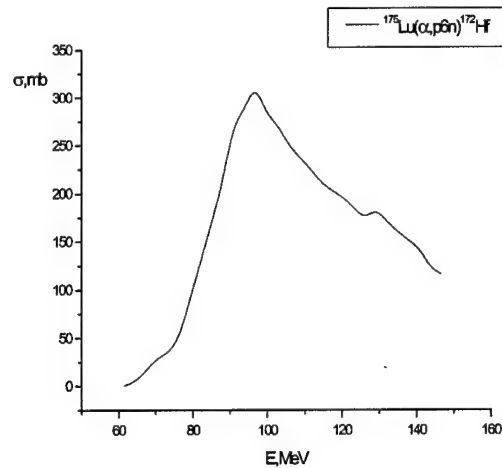
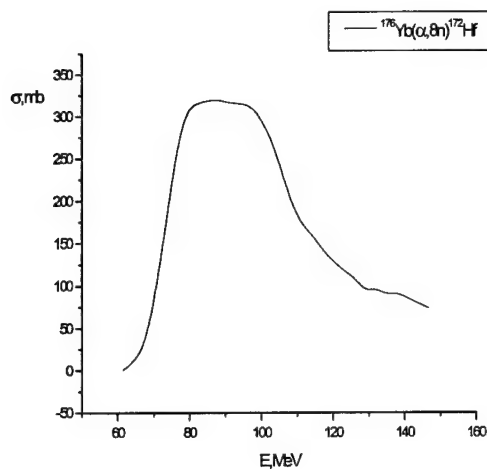
## 5. STAPRE and ALICE code simulations for cross-sections of the most undesirable isotopes accumulated in the spallation reactions

If we consider the question of quality for the produced  $^{178\text{m}}\text{Hf}$  isomeric sources, then one should take into account the production of other Hf isotopes, at least radioactive and rather long-lived ones. Fortunately, there is only one Hf isotope, half-life of which is comparable with the half-life of  $^{178\text{m}}\text{Hf}$  and that is  $^{172}\text{Hf}$  ( $T_{1/2} = 1.87$  years). This isotope would be the most undesirable admixture for all new samples. The ration  $^{178\text{m}}\text{Hf}/^{172}\text{Hf}$  in the sample seems to be useful for estimating of cooling times when any prediscovey experiments can be started.

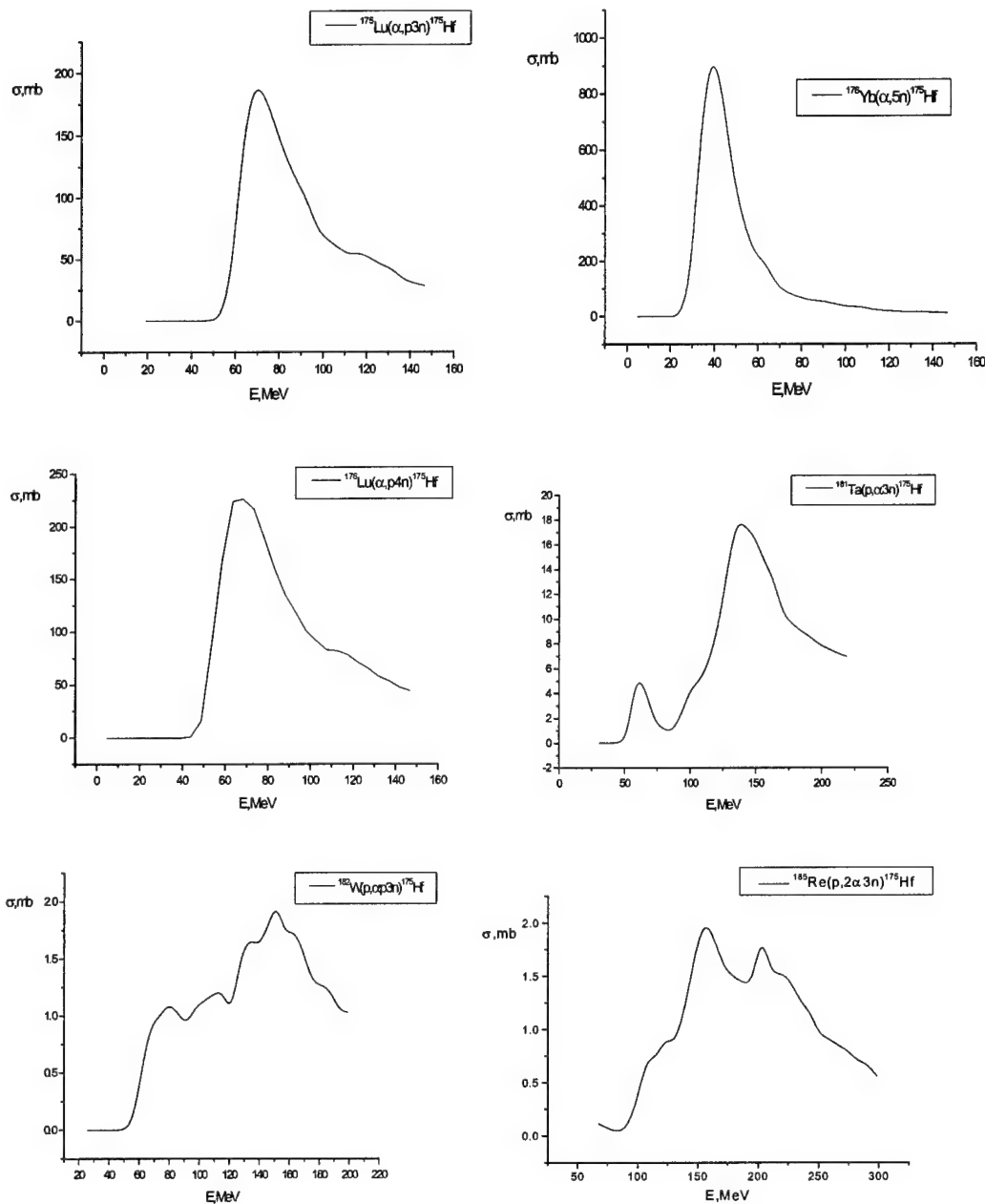
In the case of  $\alpha$ -particles the production of this most undesirable Hf isotope can be significantly reduced by choosing the energy range of bombarding  $\alpha$ -particles less than at least 80 MeV, since the cross-section reaches maximum at this energy for  $^{176}\text{Yb}$  (for  $^{175}\text{Lu}$  and  $^{176}\text{Lu}$  - almost at 100 MeV).

In the event of protons with energy around 100 MeV the production of  $^{172}\text{Hf}$  seems to be high, since the cross-section reaches maximum at about 80 MeV for  $^{181}\text{Ta}$  (and the local

maximums at about 100 MeV for  $^{185}\text{Re}$  and  $^{182}\text{W}$ ). At the same time, in the case of  $^{182}\text{W}$  and  $^{185}\text{Re}$ , the production of  $^{172}\text{Hf}$  is much lower, than in the event of  $^{181}\text{Ta}$ , but one should not forget that  $^{178\text{m}}\text{Hf}$  isomers are produced in much less quantities as well.

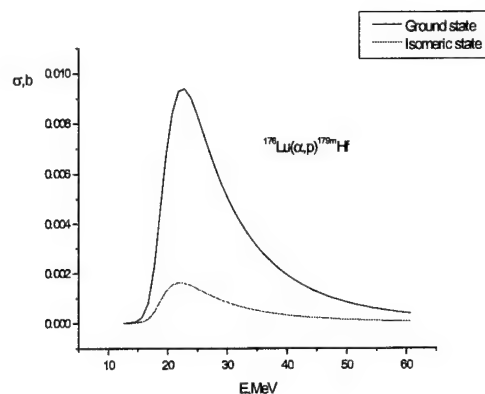
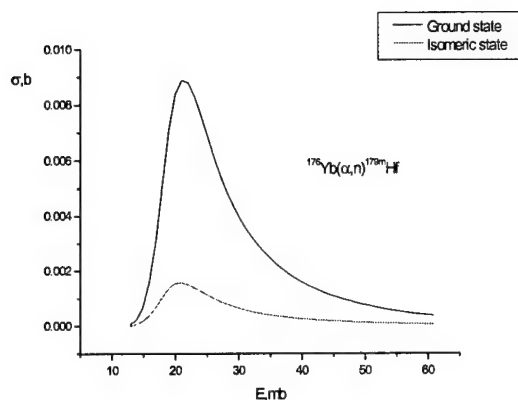


Other Hf isotopes could be a problem for the production of  $^{178m}\text{Hf}$  isomeric sources as well. The most undesirable Hf isotopes, save for already discussed  $^{172}\text{Hf}$ , turn out to be  $^{175}\text{Hf}$ ,  $^{179m}\text{Hf}$ ,  $^{181}\text{Hf}$ .



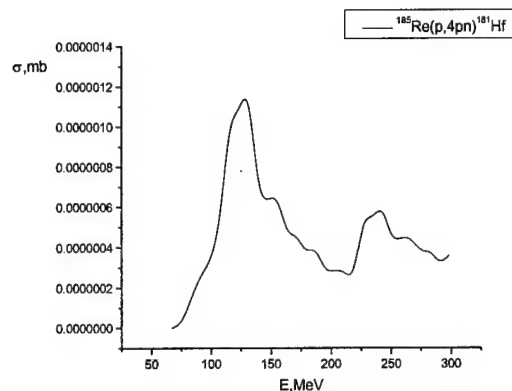
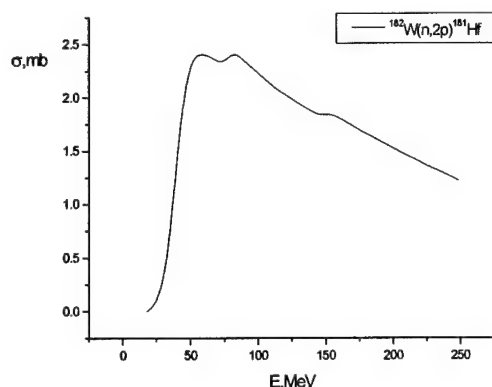
So, for  $^{175}\text{Hf}$  the situation looks almost the same as for  $^{172}\text{Hf}$ . Really, in the case of  $^{176}\text{Yb}$  the maximum cross-section is reached at lower energy, than in the event of  $^{175}\text{Lu}$  and  $^{176}\text{Lu}$ , and maximal value is around 4-5 times higher. For protons the situation repeats absolutely the one for  $^{172}\text{Hf}$ .



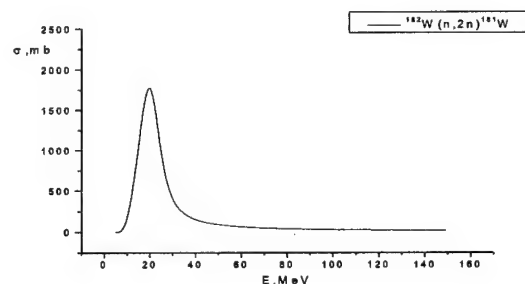
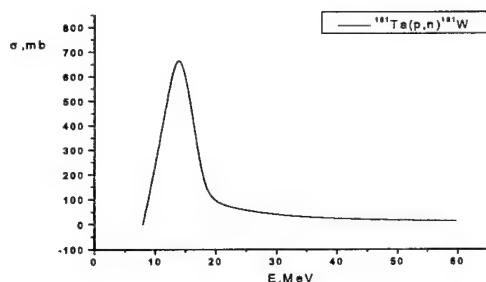


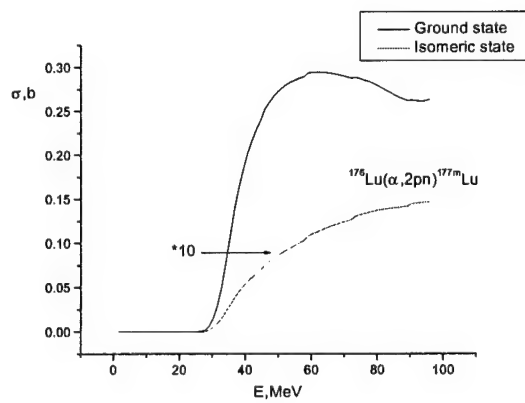
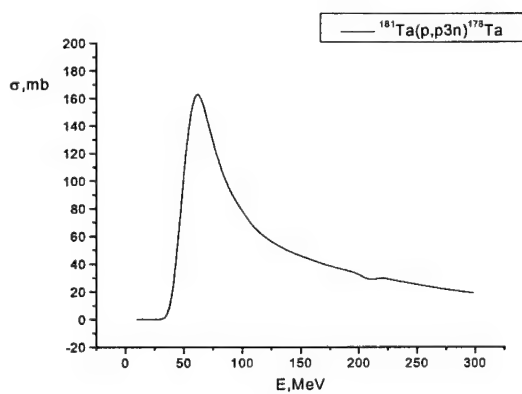
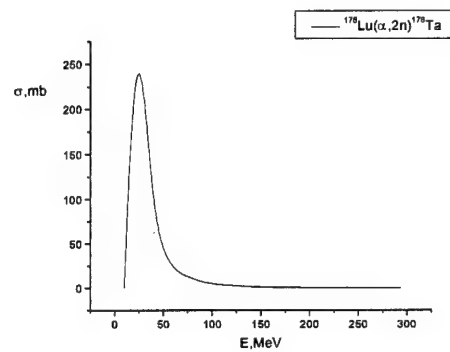
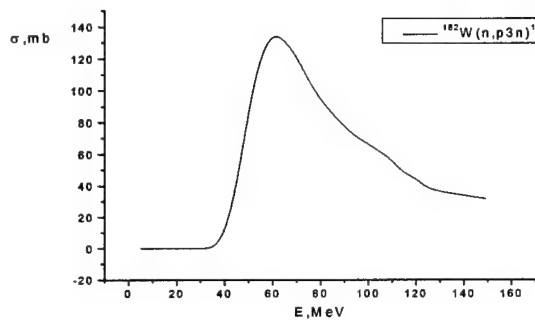
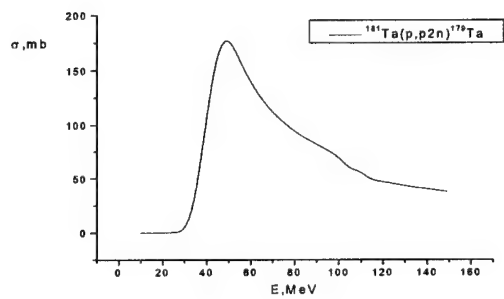
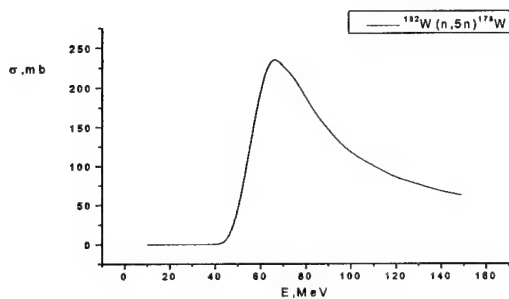
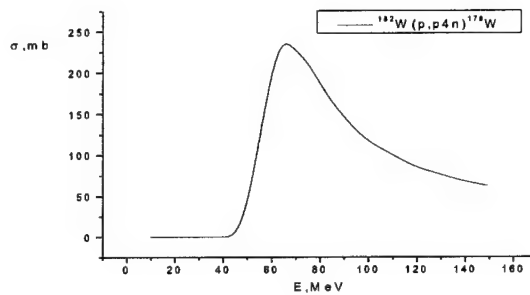
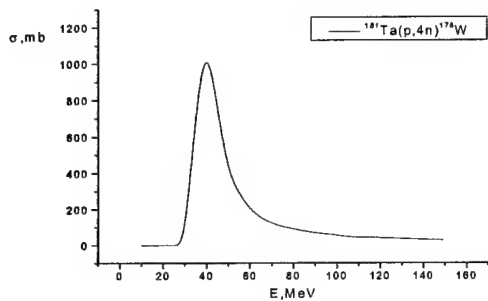
The behavior of excitation functions for  $^{179m}\text{Hf}$  turns out to be practically the same for both nuclear reactions with  $\alpha$ -particles, in which it could be produced.

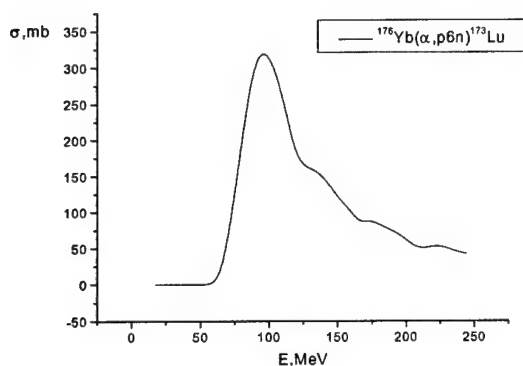
For two nuclear reactions taken as an example, the behavior of excitation functions for  $^{181}\text{Hf}$  looks as follows:



Another factor that influences quite significantly upon the quality of produced  $^{178m}\text{Hf}$  isomeric sources is the production of other long-lived radionuclides. So we have also calculated a number of such undesired by-products or admixtures, which could be accumulated in targets during the irradiation, such as for instance  $^{181}\text{W}$ ,  $^{178}\text{W}$ ,  $^{182}\text{Ta}$ ,  $^{179}\text{Ta}$ ,  $^{178}\text{Ta}$ ,  $^{177m}\text{Lu}$ ,  $^{174m,g}\text{Lu}$ ,  $^{173}\text{Lu}$ ,  $^{169}\text{Yb}$ ,  $^{171}\text{Tm}$ ,  $^{170}\text{Tm}$ ,  $^{168}\text{Tm}$ . The excitation functions for some of these long-lived radionuclides, for which cross-sections reach 100 mb or more, are presented below.







Fortunately, any long-lived admixtures, save for Hf isotopes, can be eliminated by radio-chemical methods, though their concentrations in produced raw material could also be important due to chemical processing problems of Hf fraction separation from the targets after their irradiation and the situation looks as follows. In the case of  $\alpha$ -particles only when Lu targets are used, there could be a difficulty of Ta fraction isolation from the Hf one. All the other undesired admixtures turn to be a number of rare-earth elements, the isolation of which from Hf fraction is not be a problem at all.

In the case of protons and neutrons and depending on the particular nuclear reaction under discussion, one should solve the problem of separation for Ta, W, Re or even Os fractions as well. At the same time, such methods and techniques have been well developed and tested in many experiments. Thus, first of all the target (W or Ta or Re) used to be dissolved in concentrated hydrofluoric acid with addition of  $\text{HNO}_3$ . Then, Hf fraction is isolated from W, Ta, Re and Os ones and from other radionuclides, the most part of which are rare-earth elements. And at last, Hf fraction is fine rectified from the remaining W, Ta, Re and Os admixtures.

As one can notice, the production of  $^{178}\text{W}$ , just as  $^{178}\text{Ta}$  and  $^{179}\text{Ta}$ , happens to be very high in all the nuclear reactions with protons and neutrons (only  $^{178}\text{Ta}$  and  $^{179}\text{Ta}$  in the nuclear reactions with  $\alpha$ -particles and when Lu targets are used). It means that for the production of high quality  $^{178\text{m}2}\text{Hf}$  isomeric sources one should choose the irradiation mode with optimal time of irradiation, otherwise the concentration of  $^{178}\text{Hf}$  and  $^{179}\text{Hf}$  ground states would be very high.

## 6. Selection of spallation reactions for optimal production of $^{178\text{m}2}\text{Hf}$ isomer

Using the results of our calculations one can estimate at least qualitatively the optimal irradiation mode and cost of  $^{178\text{m}}\text{Hf}$  isomeric source production, just as all the parameters of produced isomeric sources. However, in order to do it quantitatively one needs to analyze in detail quite a lot of data and perform a comparative analysis for nuclear reactions under consideration.

Looking at the results of such comparative analysis for isotopes yield (nuclei per incident particle) in the case of nuclear reactions with  $\alpha$ -particles (Table 2) one can see that, though the attractiveness of  $^{176}\text{Lu}(\alpha, pn)^{178\text{m}}\text{Hf}$  reaction is rather poor,  $^{175}\text{Lu}(\alpha, p)^{178\text{m}}\text{Hf}$  and  $^{176}\text{Yb}(\alpha, 2n)^{178\text{m}}\text{Hf}$  reactions look quite competitive. Really, depending on the energy of  $\alpha$ -particles the productivity of  $^{176}\text{Yb}(\alpha, 2n)^{178\text{m}}\text{Hf}$  reaction is only 1.7 - 1.9 times higher, than the productivity of  $^{175}\text{Lu}(\alpha, p)^{178\text{m}}\text{Hf}$  one. At the same time, first, the yield of  $^{178\text{g}}\text{Hf}$  ground state turns out to be 12.2 - 12.0 times higher as well. Secondly, the yield of most undesirable admixture  $^{172}\text{Hf}$  would be 1 - 1.6 times higher also. Thirdly, the yield of another long-lived radioactive Hf isotope, such as  $^{175}\text{Hf}$  turns out to be 4.9 - 2.4 times higher as well.

Table 2. Isotope yields (nuclei per incident particle) in the case of nuclear reactions with  $\alpha$ -particles.

Product	E $_{\alpha}$ , MeV						
	40	50	60	70	80	90	100
<b><math>^{175}\text{Lu}</math></b>							
$^{172}\text{Hf}$	0	0	1.768E-8	6.385E-6	4.504E-5	1.839E-4	4.433E-4
$^{174}\text{Hf}$	1.265E-11	4.572E-6	7.971E-5	2.553E-4	4.517E-4	6.137E-4	7.456E-4
$^{175}\text{Hf}$	5.777E-7	4.47E-5	1.533E-4	2.527E-4	3.428E-4	4.178E-4	4.782E-4
$^{176}\text{Hf}$	2.722E-5	1.03E-4	1.651E-4	2.238E-4	2.817E-4	3.278E-4	3.61E-4
$^{177}\text{Hf}$	3.146E-5	5.542E-5	7.576E-5	9.308E-5	1.077E-4	1.198E-4	1.307E-4
$^{178g}\text{Hf}$	4.317E-6	4.865E-6	5.146E-6	5.295E-6	5.374E-6	5.423E-6	5.467E-6
$^{178m}\text{Hf}$	9.217E-7	1.035E-6	1.092E-6	1.12E-6	1.134E-6	1.142E-6	1.15E-6
$\Sigma\text{Hf}$	6.44964E-05	0.000214	0.0004801	0.0008377	0.001235	0.00167	0.002165
$^{178m}\text{Hf}/\Sigma\text{Hf}$	0.014290717	0.004846	0.0022744	0.001337	0.000918	0.000684	0.000531
<b><math>^{176}\text{Lu}</math></b>							
$^{172}\text{Hf}$	0	0	2.561E-13	2.096E-8	2.992E-6	3.708E-5	1.783E-4
$^{174}\text{Hf}$	0	3.6E-8	8.87E-6	7.727E-5	2.432E-4	4.608E-4	6.546E-4
$^{175}\text{Hf}$	1.114E-10	4.577E-6	7.266E-5	2.096E-4	3.552E-4	4.786E-4	5.73E-4
$^{176}\text{Hf}$	2.833E-6	5.405E-5	1.596E-4	2.746E-4	3.822E-4	4.681E-4	5.287E-4
$^{177}\text{Hf}$	2.636E-5	9.045E-5	1.484E-4	1.951E-4	2.372E-4	2.726E-4	3.043E-4
$^{178g}\text{Hf}$	2.073E-5	2.541E-5	2.821E-5	3.017E-5	3.167E-5	3.294E-5	3.422E-5
$^{178m}\text{Hf}$	9.145E-8	1.164E-7	1.344E-7	1.514E-7	1.692E-7	1.886E-7	2.104E-7
$^{179g}\text{Hf}$	3.873E-6	4.377E-6	4.631E-6	4.758E-6	4.821E-6	4.854E-6	4.885E-6
$^{179m}\text{Hf}$	7.913E-7	8.876E-7	9.36E-7	9.601E-7	9.718E-7	9.781E-7	9.84E-7
$\Sigma\text{Hf}$	5.46789E-05	0.00018	0.0004234	0.0007926	0.001258	0.001756	0.002279
$^{178m}\text{Hf}/\Sigma\text{Hf}$	0.001672493	0.000647	0.0003174	0.000191	0.000134	0.000107	9.23E-05
<b><math>^{176}\text{Yb}</math></b>							
$^{172}\text{Hf}$	0	0	1.689E-8	1.663E-5	1.84E-4	4.302E-4	7.059E-4
$^{174}\text{Hf}$	0	2.075E-5	3.206E-4	7.849E-4	1.1E-3	1.31E-3	1.46E-3
$^{175}\text{Hf}$	2.823E-6	2.763E-4	6.663E-4	8.662E-4	9.892E-4	1.09E-3	1.16E-3
$^{176}\text{Hf}$	3.315E-4	7.526E-4	9.528E-4	1.05E-3	1.12E-3	1.19E-3	1.2410-3
$^{177}\text{Hf}$	4.392E-4	5.461E-4	6.053E-4	6.467E-4	6.762E-4	6.983E-4	7.15E-4
$^{178g}\text{Hf}$	5.259E-5	5.732E-5	6.023E-5	6.221E-5	6.365E-5	6.474E-5	6.564E-5
$^{178m}\text{Hf}$	1.597E-6	1.749E-6	1.858E-6	1.946E-6	2.023E-6	2.093E-6	2.158E-6
$^{179g}\text{Hf}$	3.468E-6	3.93E-6	4.187E-6	4.34E-6	4.429E-6	4.476E-6	4.508E-6
$^{179m}\text{Hf}$	9.766E-7	1.102E-6	1.172E-6	1.214E-6	1.238E-6	1.251E-6	1.26E-6
$\Sigma\text{Hf}$	0.000832155	0.00166	0.0026125	0.0034341	0.004141	0.004791	0.004114
$^{178m}\text{Hf}/\Sigma\text{Hf}$	0.001919115	0.001054	0.0007112	0.0005667	0.000489	0.000437	0.000524

And at last, the yields of stable Hf isotopes, such as  $^{174}\text{Hf}$ ,  $^{176}\text{Hf}$  and  $^{177}\text{Hf}$  would to be 0.5 – 2.0, 12.2 – 3.4 and 14.0 – 5.5 times higher, respectively. As a result, in the case of  $^{175}\text{Lu}$  ( $\alpha$ , p)  $^{178m}\text{Hf}$  reaction the concentration of  $^{178m}\text{Hf}$  isomer in Hf fraction of the target happens to be higher over all the energy range and at least for some applications  $^{175}\text{Lu}$  ( $\alpha$ , p)  $^{178m}\text{Hf}$  reaction can be more preferable, than  $^{176}\text{Yb}$  ( $\alpha$ , 2n)  $^{178m}\text{Hf}$  one. Moreover, the energy dependence of  $^{178m}\text{Hf}$  isomer concentration in Hf fraction of the target for these two reactions looks rather different (in the event of  $^{176}\text{Yb}$  ( $\alpha$ , 2n)  $^{178m}\text{Hf}$  there is a local minimum at 90 keV).

Looking at the results of such comparative analysis for isotopes yield in the case of nuclear reactions with protons (Table 3) one can see that the attractiveness of  $^{185}\text{Re}$  (p, 2 $\alpha$ )  $^{178m}\text{Hf}$  and  $^{182}\text{W}$  (p,  $\alpha$ p)  $^{178m}\text{Hf}$  reactions is rather low compared to  $^{181}\text{Ta}$  (p,  $\alpha$ )  $^{178m}\text{Hf}$  one, though at the same time  $^{185}\text{Re}$  (p, 2 $\alpha$ )  $^{178m}\text{Hf}$  and  $^{182}\text{W}$  (p,  $\alpha$ p)  $^{178m}\text{Hf}$  reactions look rather competitive. Let's compare in detail two nuclear reactions, for instant,  $^{181}\text{Ta}$  (p,  $\alpha$ )  $^{178m}\text{Hf}$  and  $^{182}\text{W}$  (p,  $\alpha$ p)  $^{178m}\text{Hf}$ . As a result, one can see that depending on the energy of protons the productivity of  $^{181}\text{Ta}$  (p,  $\alpha$ )  $^{178m}\text{Hf}$  reaction is more than three orders of magnitude higher,

Table 3. Isotope yields (nuclei per incident particle) in the case of nuclear reactions with protons.

Product	20	30	40	50	60	70	80	90	100
<b><sup>181</sup>Ta</b>									
<sup>180</sup> Hf	3.260E-9	1.024E-6	1.151E-5	4.624E-5	1.006E-4	1.654E-4	2.371E-4	3.123E-4	3.929E-4
<sup>179</sup> Hf	0.000	0.000	8.295E-9	5.559E-6	4.319E-5	1.012E-4	1.595E-4	2.199E-4	2.787E-4
<sup>178</sup> Hf	7.072E-9	2.061E-8	2.101E-8	6.787E-8	6.755E-6	5.959E-5	1.611E-4	2.721E-4	3.921E-4
<sup>177</sup> Hf	2.328E-9	3.155E-7	7.553E-7	8.396E-7	1.238E-6	5.780E-6	4.462E-5	1.429E-4	2.533E-4
<sup>176</sup> Hf	0.000	9.271E-8	4.523E-6	8.742E-6	1.035E-5	1.359E-5	2.754E-5	7.190E-5	1.878E-4
<sup>175</sup> Hf	0.000	0.000	1.752E-6	2.543E-5	4.413E-5	5.238E-5	6.367E-5	9.682E-5	1.454E-4
<sup>174</sup> Hf	0.000	0.000	1.391E-8	1.222E-5	9.380E-5	1.750E-4	2.248E-4	2.707E-4	3.592E-4
<sup>172</sup> Hf	0.000	0.000	0.000	0.000	7.090E-8	3.239E-5	2.241E-4	4.896E-4	6.821E-4
<sup>178m</sup> Hf	9.004E-11	3.220E-10	3.301E-10	1.220E-9	1.411E-7	1.414E-6	4.529E-6	8.700E-6	1.413E-5
$\Sigma$ Hf	1.275E-08	1.45E-06	1.858E-05	9.91E-05	0.0003	0.000607	0.001147	0.001885	0.002706
<sup>178m</sup> Hf/ $\Sigma$ Hf	0.007062	0.000222	1.776E-05	1.231E-05	0.00047	0.00233	0.003949	0.004616	0.005222
<sup>178</sup> Ta	0.000	8.640E-12	3.944E-9	3.515E-7	1.275E-6	2.374E-6	3.341E-6	4.182E-6	4.941E-6
<sup>179</sup> Ta	2.274E-12	1.960E-9	2.259E-7	1.097E-6	2.063E-6	2.915E-6	3.694E-6	4.422E-6	5.106E-6
<sup>178</sup> W	0.000	1.151E-8	3.322E-6	7.103E-6	8.843E-6	9.902E-6	1.070E-5	1.135E-5	1.191E-5
<b><sup>182</sup>W</b>									
<sup>180</sup> Hf	0.000	0.000	0.000	5.024E-14	6.697E-12	1.295E-10	3.839E-10	6.742E-10	9.168E-10
<sup>179</sup> Hf	0.000	0.000	0.000	6.839E-14	4.840E-11	4.728E-10	2.510E-9	6.613E-9	1.112E-8
<sup>178</sup> Hf	0.000	8.683E-12	1.293E-9	5.291E-9	8.503E-9	1.476E-8	3.921E-8	8.815E-8	1.921E-7
<sup>177</sup> Hf	0.000	0.000	2.184E-9	6.091E-8	1.895E-7	3.162E-7	4.708E-7	7.675E-7	1.222E-6
<sup>176</sup> Hf	0.000	0.000	8.458E-11	8.695E-8	1.045E-6	2.768E-6	4.453E-6	6.217E-6	8.803E-6
<sup>175</sup> Hf	0.000	0.000	0.000	2.179E-9	7.809E-7	5.951E-6	1.403E-5	2.267E-5	3.193E-5
<sup>174</sup> Hf	0.000	0.000	0.000	0.000	4.947E-8	3.911E-6	2.506E-5	5.935E-5	9.830E-5
<sup>172</sup> Hf	0.000	0.000	0.000	0.000	0.000	1.019E-10	4.050E-7	1.504E-5	9.404E-5
<sup>178m</sup> Hf	0.000	3.286E-13	2.217E-11	1.072E-10	1.855E-10	3.572E-10	1.295E-9	3.563E-9	6.178E-9
$\Sigma$ Hf	0.000	9.01E-12	3.58E-09	1.55E-07	2.07E-06	1.3E-05	4.45E-05	0.000104	0.000235
<sup>178m</sup> Hf/ $\Sigma$ Hf	0.000	0.036464	0.006186	0.00069	8.92E-05	2.76E-05	2.91E-05	3.42E-05	2.63E-05
<sup>178</sup> Ta	1.108E-12	7.728E-10	1.591E-9	1.783E-9	4.525E-9	2.035E-8	8.494E-8	2.156E-7	3.686E-7
<sup>179</sup> Ta	2.120E-11	6.764E-11	6.787E-11	2.633E-10	1.009E-8	7.959E-8	2.014E-7	3.256E-7	4.418E-7
<sup>178</sup> W	0.000	0.000	1.728E-11	6.861E-8	8.636E-7	2.444E-6	4.020E-6	5.372E-6	6.526E-6
<b><sup>185</sup>Re</b>									
<sup>180</sup> Hf	0.000	0.000	0.000	5.440E-15	1.528E-12	5.628E-11	2.657E-10	5.682E-10	1.177E-9
<sup>179</sup> Hf	0.000	0.000	0.000	5.151E-14	2.486E-11	2.114E-10	1.106E-9	4.330E-9	9.689E-9
<sup>178</sup> Hf	0.000	2.842E-14	3.141E-12	4.576E-12	3.154E-11	2.953E-9	1.799E-8	4.865E-8	1.116E-7
<sup>177</sup> Hf	0.000	0.000	5.355E-11	1.853E-9	4.304E-9	6.256E-9	4.504E-8	2.494E-7	5.253E-7
<sup>176</sup> Hf	0.000	0.000	4.093E-12	6.508E-9	7.605E-8	1.427E-7	1.869E-7	5.522E-7	1.841E-6
<sup>175</sup> Hf	0.000	0.000	0.000	2.722E-10	8.525E-8	6.601E-7	1.230E-6	1.584E-6	3.102E-6
<sup>174</sup> Hf	0.000	0.000	0.000	0.000	3.744E-9	6.159E-7	3.855E-6	7.584E-6	1.008E-5
<sup>172</sup> Hf	0.000	0.000	0.000	0.000	0.000	3.638E-12	4.774E-8	3.835E-6	1.901E-5
<sup>178m</sup> Hf	0.000	2.220E-16	1.310E-14	2.398E-14	3.395E-13	4.499E-11	3.161E-10	9.381E-10	2.345E-9
$\Sigma$ Hf	0.000	2.86E-14	6.08E-11	8.64E-09	1.69E-07	1.43E-06	5.38E-06	1.39E-05	3.47E-05
<sup>178m</sup> Hf/ $\Sigma$ Hf	0.000	0.007751	0.000215	2.78E-06	2E-06	3.15E-05	5.87E-05	6.77E-05	6.76E-05
<sup>178</sup> Ta	0.000	0.000	7.105E-15	7.48E-12	1.077E-9	6.885E-9	1.745E-8	2.909E-8	4.122E-8
<sup>179</sup> Ta	0.000	0.000	7.105E-15	7.482E-12	1.077E-9	6.885E-9	1.745E-8	2.909E-8	4.122E-8
<sup>178</sup> W	0.000	0.000	5.159E-11	1.672E-8	1.036E-7	2.050E-7	2.602E-7	3.098E-7	3.929E-7

than the productivity of  $^{182}\text{W}(\text{p}, \alpha\text{p})^{178\text{m}}\text{Hf}$  one. At the same time, the yield of  $^{178\text{g}}\text{Hf}$  ground state turns out to be proportionally higher as well. On the other hand, though for 80 MeV protons the yield of most undesirable admixture  $^{172}\text{Hf}$  would be almost proportionally higher also, however for 100 MeV protons its yield turns out to be only 7.3 times higher. Moreover, the yield of another long-lived radioactive Hf isotope, such as  $^{175}\text{Hf}$  would be only 4.5 and 46 times higher at the energy of protons 80 and 100 MeV, respectively as well. The yields of stable Hf isotopes, such as  $^{174}\text{Hf}$ ,  $^{176}\text{Hf}$ ,  $^{177}\text{Hf}$ ,  $^{179}\text{Hf}$  and  $^{180}\text{Hf}$  turn out to be 9.0 and 3.7, 6.2 and  $2.1 \times 10^2$ , 95 and  $2.1 \times 10^2$ ,  $6.4 \times 10^4$  and  $2.5 \times 10^4$ ,  $6.2 \times 10^5$  and  $4.3 \times 10^5$  times higher at the energy of protons 80 and 100 MeV, respectively. And at last, the yields of most undesirable radioactive isotopes different from Hf, such as  $^{178}\text{W}$ ,  $^{178}\text{Ta}$  and  $^{179}\text{Ta}$  would be only 2.7 and 1.8, 39.3 and 13.4, 18.3 and 11.6 times higher at the energy of protons 80 and 100 MeV, respectively. Thus,  $^{181}\text{Ta}(\text{p}, \alpha)^{178\text{m}}\text{Hf}$  reaction looks still more attractive from many points of view. Really, the most vital factor is much higher productivity of  $^{181}\text{Ta}(\text{p}, \alpha)^{178\text{m}}\text{Hf}$  reaction compared to other nuclear reactions with protons. Another essential advantage of this reaction at the energy of protons 100 MeV is non-proportionally low yield of  $^{172}\text{Hf}$ . As a result, the concentration of  $^{178\text{m}}\text{Hf}$  isomer in Hf fraction of the target turns out to be higher for the nuclear reaction with Ta target.

If we compare the nuclear reaction with  $\alpha$ -particles,  $^{176}\text{Yb}(\alpha, 2\text{n})^{178\text{m}}\text{Hf}$  and the nuclear reaction with protons,  $^{181}\text{Ta}(\text{p}, \alpha)^{178\text{m}}\text{Hf}$  one can note that at the same energy of  $\alpha$ -particles and protons of 100 MeV the productivity of  $^{181}\text{Ta}(\text{p}, \alpha)^{178\text{m}}\text{Hf}$  reaction is 6.5 times higher than the productivity of  $^{176}\text{Yb}(\alpha, 2\text{n})^{178\text{m}}\text{Hf}$  one. At the same time, the yield of  $^{178\text{g}}\text{Hf}$  ground state turns out to be 6.0 times higher as well, though the yield of most undesirable admixture  $^{172}\text{Hf}$  seems to be practically the same. On the other hand, the yield of another long-lived radioactive Hf isotope, such as  $^{175}\text{Hf}$  turns out to be 275 times lower. The yields of stable Hf isotopes, such as  $^{174}\text{Hf}$ ,  $^{176}\text{Hf}$ ,  $^{177}\text{Hf}$  and  $^{179}\text{Hf}$  turn out to be 4.1, 6.6 and 2.8 times lower and 12.3 times higher, respectively. One can note that the concentration of stable Hf isotopes in Hf fraction of the target appears to be practically the same for both nuclear reactions, though the concentration of  $^{172}\text{Hf}$  and  $^{175}\text{Hf}$  should be non-proportionally lower. As a result, in the event of  $^{181}\text{Ta}(\text{p}, \alpha)^{178\text{m}}\text{Hf}$  the concentration of  $^{178\text{m}}\text{Hf}$  isomer in Hf fraction of the target appears to be an order of magnitude higher, than in the event of  $^{176}\text{Yb}(\alpha, 2\text{n})^{178\text{m}}\text{Hf}$ , and has a tendency to increase with energy.

Thus, nuclear reactions with  $\alpha$ -particles could be preferable only when one needs to produce rather small, but of maximal quality,  $^{178\text{m}}\text{Hf}$  sources. For instant, in the case of  $^{175}\text{Lu}(\alpha, \text{p})^{178\text{m}}\text{Hf}$  and  $^{176}\text{Yb}(\alpha, 2\text{n})^{178\text{m}}\text{Hf}$  reactions and at the energy of  $\alpha$ -particles about 40 MeV, first, no  $^{172}\text{Hf}$  would be produced at all. Secondly, in the event of  $^{175}\text{Lu}(\alpha, \text{p})^{178\text{m}}\text{Hf}$  reaction and at the energy of  $\alpha$ -particles about 40 MeV,  $^{178\text{m}}\text{Hf}$  isomer concentration in Hf fraction of the target turns out to be around 3 times higher, than in the event of  $^{181}\text{Ta}(\text{p}, \alpha)^{178\text{m}}\text{Hf}$  reaction and at the energy of protons about 100 MeV. At the same time, in the case of  $^{176}\text{Yb}(\alpha, 2\text{n})^{178\text{m}}\text{Hf}$  reaction and at the energy of  $\alpha$ -particles about 40 MeV,  $^{178\text{m}}\text{Hf}$  isomer concentration in Hf fraction of the target turns out to be around 2 times lower, than in the event of  $^{181}\text{Ta}(\text{p}, \alpha)^{178\text{m}}\text{Hf}$  reaction and at the energy of protons about 100 MeV.

## 7. Summary

$^{178\text{m}2}\text{Hf}$  isomer production in different spallation reactions with neutrons, protons and alpha-particles at projectile energies up to 100 MeV has been analyzed exploiting the STAPRE and ALICE code simulations. A number of spallation reactions has been compared taking into account not only the  $^{178\text{m}2}\text{Hf}$  isomer productivity, but first the isomeric/ground state ratios calculated by the STAPRE code, secondly, the accumulation of the most



undesirable Hf isotopes, such as  $^{172}\text{Hf}$  and  $^{175}\text{Hf}$ , thirdly, other admixtures and by-products that could degrade the quality of produced  $^{178\text{m}2}\text{Hf}$  isomer sources, including all the stable Hf isotopes as well. Possibilities and ways of the optimization for  $^{178\text{m}2}\text{Hf}$  isomer production in spallation reactions at projectile energies up to 100 MeV have been discussed and it can be considered as very important preliminary stage in order to accumulate such exotic nuclear isomers in lab-sized quantities for the reasonable costs.

The most remarkable results of our calculations are:

1. In the case of nuclear reactions with  $\alpha$ -particles, from the point of view of productivity  $^{176}\text{Yb} (\alpha, 2n) ^{178\text{m}}\text{Hf}$  reaction looks as the most attractive one, though  $^{175}\text{Lu} (\alpha, p) ^{178\text{m}}\text{Hf}$  and  $^{176}\text{Lu} (\alpha, pn) ^{178\text{m}}\text{Hf}$  are about one order of magnitude and less than two orders of magnitude worse, respectively. In the event of nuclear reactions with protons,  $^{181}\text{Ta} (p, \alpha) ^{178\text{m}}\text{Hf}$  reaction looks as the most attractive one, moreover  $^{182}\text{W} (p, \alpha p) ^{178\text{m}}\text{Hf}$  and  $^{185}\text{Re} (p, 2\alpha) ^{178\text{m}}\text{Hf}$  are less and more than three orders of magnitude worse, respectively. Furthermore, the productivity of  $^{176}\text{Yb} (\alpha, 2n) ^{178\text{m}}\text{Hf}$  reaction appears to be less than order of magnitude lower, than the productivity of  $^{181}\text{Ta} (p, \alpha) ^{178\text{m}}\text{Hf}$  one.
2. For the applications, when quite small  $^{178\text{m}}\text{Hf}$  isomer sources are required, one can produce very qualitative isomeric sources using the nuclear reactions with  $\alpha$ -particles. Furthermore, from the point of view of the  $^{178\text{m}}\text{Hf}$  isomer concentration in Hf fraction of the target, at 40 MeV  $\alpha$ -particles  $^{175}\text{Lu} (\alpha, p) ^{178\text{m}}\text{Hf}$  reaction appears to be almost an order of magnitude better, than  $^{176}\text{Yb} (\alpha, 2n) ^{178\text{m}}\text{Hf}$  reaction, although the productivity of  $^{176}\text{Yb} (\alpha, 2n) ^{178\text{m}}\text{Hf}$  reaction is still a little less than two-fold higher.

Unfortunately, it looks practically impossible to make any universal recommendations and common conclusions how to perform the optimization of  $^{178\text{m}2}\text{Hf}$  isomer production in any particular case. Really, for some prediscvery experiments even two-fold decrease of any undesirable admixture or by-product (including as  $^{172}\text{Hf}$  and  $^{175}\text{Hf}$  as all the stable Hf isotopes) could be extremely crucial, so such questions should be solved as appropriate.

As a result of our research, one can at least select, taking into account as the available nuclear facilities as the accepted costs, the most suitable nuclear reaction needed to produce significant quantities of  $^{178\text{m}2}\text{Hf}$  isomers of rather predicted quality. And although we did calibrate our calculations with the STAPRE and ALICE codes using the latest experimental data, there is a feeling that around 100 MeV the STAPRE calculations could still be incorrect even up to the order of magnitude, since along with the pre-equilibrium and evaporation models, the intranuclear cascade model of nuclear reaction shall also be used at such projectile energies. So, it would be very important to:

1. conduct the calculations using some code (or codes) valid for the spallation region, for instant any codes exploiting GIANT 4 libraries;
2. check the calculations experimentally, at least at optimal bombarding particle energies;
3. verify the results obtained for  $^{182}\text{W} (n, \alpha n) ^{178\text{m}2}\text{Hf}$  reaction both theoretically and experimentally, especially paying attention that the results of our calculations contradict significantly to the experimental data for nuclear reactions with reactor neutrons obtained recently for a number of targets of highly enriched Hf isotopes. As a good fortune,  $^{183}\text{W}$  and  $^{184}\text{W}$  are the stable isotopes. So irradiating the highly enriched  $^{182}\text{W}$  sample by reactor neutrons, one can perform very sensitive test.



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